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Dynamic Effects of Diabatization in Distillation Columns

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Abstract: The dynamic effects of diabatization in distillation columns are investigated in simulation with primary focus on the heat-integrated distillation column (HIDiC). A generic, dynamic, first-principle model has been formulated, which is flexible to describe various diabatic distillation configurations. Dynamic Relative Gain Array and Singular Value Analysis have been applied in a comparative study of a conventional distillation column and a HIDiC. The study showed increased input-output coupling due to diabatization. Feasible SISO control structures for the HIDiC were also found. Control-loop feasibility was demonstrated.

Keywords: Fluid separations, Distillation columns, Diabatic distillation, Dynamic modeling

1. INTRODUCTION

Multi-stage distillation is the most widely used industrial technique for continuously separating liquid mixtures. At the same time it is an energy intensive operation, so the technology still receives attention due to increased focus on environmental issues and resource management. In conventional distillation columns (CDiC) heat is added at the highest temperature and heat is removed at the lowest temperature, thereby preventing integration of the utility streams from the condenser and reboiler (Jana, 2010). Exergy (available energy) is thus said to be degraded throughout the distillation column. A Second-Law analysis (Fitzmorris and Mah, 1980) can quantify this exergy degradation. Such analysis reveals that industrial distillation columns operate at Second-Law efficiencies in the range of 5-20% (de Koeijer and Kjelstrup, 2000). These low Second-Law efficiencies indicate a substantial potential for improvements.

Various alternative distillation column configurations with higher Second-Law efficiencies have been suggested. One group of alternative configurations are the diabatic distillation columns. In a diabatic distillation column the heat required to perform the separation is added and/or removed throughout the column. An example is the heat-integrated distillation column (HIDiC) which was conceptually introduced by Mah et al. (1977). An illustration of a CDiC and a general HIDiC is given in Figure 1.

Internal heat transfer in the HIDiC is realized by operating the rectifying section at higher pressure than the stripping section, using vapor recompression. This heat transfer facilitates gradual boil-up throughout the stripper and condensation throughout the rectifier leading to an improved Second-Law efficiency (Nakaiwa et al., 1998a). The HIDiC uses significantly less utility in form of steam and cooling water compared to the CDiC. Instead, energy needs to be supplied to the compressor. Olujic et al. (2003) has shown that this operation significantly reduces the operation cost of the separation.

Extensive efforts have been made to develop this technology during the past 15 years, both theoretically (e.g. Nakaiwa et al. (2003); Gadalla et al. (2007); Jana (2010)) and experimentally (Naito et al., 2000; Brunisma et al., 2012). Despite demonstrations of large energy savings of the HIDiC compared to the CDiC and manageable operability, it has not yet been accepted by the industry. This could be due to lack of mature methods for designing these more complex configurations (Olujic et al., 2006; Gadalla et al., 2007; Suphanit, 2010). Furthermore operation is more complex as a result of the high degree of process integration (Nakaiwa et al., 1998b; Zhu and Liu, 2005b).

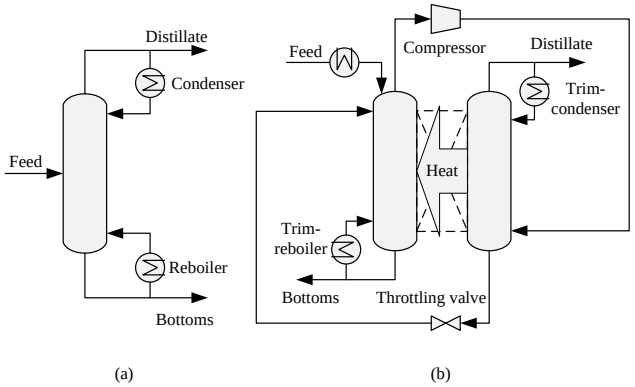

Fig. 1. Schematic representation of (a) conventional distillation column (CDiC), (b) general heat-integrated distillation column (HIDiC)

The scope of this study is to investigate the challenges related to process control which may arise from diabatization. We present a generic model for simulating operation of both the CDiC and HIDiC configurations. A SISO control structure selection is based on dynamic analysis

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tools and is verified in closed-loop simulations. The paper is organized with a presentation of the model in section 2. The simulation case is defined and the results presented in section 3 before the concluding remarks in section 4. Section 5 contains the nomenclature list.

2. DISTILLATION COLUMN MODEL

The modeling in this section forms the basis of the subsequent simulations presented in this paper. We give a detailed description of the distillation column configurations considered in this paper and their mathematical form, i.e. the dynamic model.

2.1 System Description

A distillation column consists of a rectifying section which contains the trays above the feed location and a stripping section which contains the feed tray and the trays below. In the rectifying section, the most volatile component is concentrated towards the top of the column. In the stripping section, the most volatile component is stripped off the down-flowing liquid, producing a bottom product rich in the least volatile component. A fraction of the condensed top product is returned to the column as a reflux while a fraction of the bottom product is vaporized and returned in the bottom of the column. By manipulating the amount of reflux and boilup, the purities of the top and bottom products can be adjusted. In a conventional (adiabatic) distillation column the column itself is thermally insulated from the surroundings. A diabatic distillation column on the other hand, exchanges heat with the surroundings on each tray. This can significantly reduce the need for condenser and reboiler duties. Figure 2 displays a general representation of a distillation column.

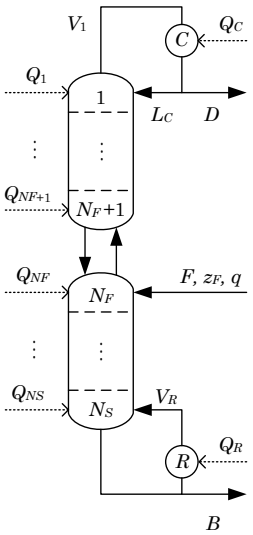

Fig. 2. General representation of a distillation column for separation of binary mixtures. The nomenclature is given in section 5

2.2 Dynamic Model

Inspired by the works of Liu and Qian (2000), Skogestad and Morari (1988), and Skogestad (1997) a dynamic model is formulated to describe both an adiabatic and a diabatic distillation column. The dynamic model includes mass and energy balances with the following assumptions:

(1) A Lewis/Randall ideal, binary mixture is considered
(2) A tray column model is used
(3) Ideal mixing and equilibrium on each tray
(4) Changes in sensible heat have been neglected
(5) An equal number of trays in the rectifying and stripping sections
(6) Vapor holdup is neglected corresponding to constant pressure in the CDiC and constant pressures in the rectifying and stripping sections in the HIDiC
(7) Liquid flows are described by linearized tray hydraulics

The model equations are summarized in Table 1 and the corresponding nomenclature list is given in section 5. As can be seen, the expression for the Q_i terms determine the configuration of the distillation column.

2.3 Control Degrees of Freedom Analysis

A crucial element in the operation of a distillation column is the selection of an efficient control structure, i.e. the pairing of controlled and manipulated variables. Control degrees of freedom analysis were performed on a CDiC and a HIDiC. A CDiC has five control degrees of freedom, namely the condenser duty, the reboiler duty, the distillate flow rate, the bottom flow rate and the reflux flow rate. Three holdups must be controlled, which are the reflux drum holdup, the reboiler holdup and the column vapor holdup, leaving the two product purities left for control. When considering the general HIDiC configuration as illustrated in Figure 1, two additional control degrees of freedom exist. These are the pressure difference between the rectifying and stripping sections and the feed preheater duty. As a result, the complexity of the control structure synthesis problem has increased, thanks to diabatization since the number of combinations have increased.

3. SIMULATION STUDY

The model described in section 2 has been implemented in MATLAB®. This section outlines the considered case study, the methods applied for analysis, and finally the results and a discussion of these. A binary, equimolar mixture of benzene-toluene is a common example mixture (Zhu and Liu, 2005a; Huang et al., 2007) and is used in this work as well.

3.1 Case-formulation

The idea is to compare the dynamics of an adiabatic distillation column (the CDiC) with a diabatic distillation column (the HIDiC), so the design degrees of freedom must be chosen according to this. The specific HIDiC configuration, presented in this paper, is the configuration as seen in Figure 1(b) without a feed preheater. The feed stream for the two configurations must obviously be the same, i.e. the feed flow rate, composition and thermal condition. The stripping section pressure of both configurations are fixed at 0.1013 MPa. The pressure of the rectifying section of the CDiC is identical to the pressure in the stripping section

while the pressure difference between the rectifying and stripping sections of the HIDiC is specified as 0.217 MPa. Additionally the product specifications are 99% benzene in the distillate and 5% benzene in the bottom product. It was furthermore decided that the CDiC and HIDiC should be designed with equal number of trays such that the hydraulic lag in the column, i.e. the time a change in the liquid flow in the top is perturbed to the bottom, is unaffected by the configuration type. A complete list of all specifications related to column design and mixture properties are given in Table 2.

Table 2. Stationary operation data and model parameters for numerical studies related to the separation of a binary mixture of benzene-toluene

General operation conditions		
Number of stages	20	-
Feed stage location	11	-
Feed flow	100	$\text{kmol} \cdot \text{h}^{-1}$
Feed benzene content	0.50	-
Feed thermal condition	1	-
Stripping section pressure	0.1013	MPa
Top purity	0.99	-
Bottom purity	0.95	-
Nominal tray holdup	0.83	kmol
CDiC specific operation conditions		
Reflux flow rate	86.6	$\text{kmol} \cdot \text{h}^{-1}$
Vapor boilup rate	134	$\text{kmol} \cdot \text{h}^{-1}$
Pressure diff. between sections	0	MPa
HIDiC specific operation conditions		
Reflux flow rate	23.9	$\text{kmol} \cdot \text{h}^{-1}$
Vapor boilup rate	71.8	$\text{kmol} \cdot \text{h}^{-1}$
Pressure diff. between sections	0.217	MPa
Model parameters		
Hydraulic time constant	0.0042	h
Heat of vaporization	30001.1	$\text{kJ} \cdot \text{kmol}^{-1}$
Relative volatility	2.317	-
Heat transfer rate	9803	$\text{kJ} \cdot \text{h}^{-1} \cdot \text{K}^{-1}$
Antoine parameters for	15.9008	-
benzene (in Torr and K)	2788.51	-
	-52.36	-

3.2 Dynamic Controllability Analysis

The control structure synthesis problem for dual composition control for a CDiC is usually solved by considering a decentralized 2×2 control problem (Skogestad et al., 1990). This procedure has been employed in this study as well, implying perfect control for the remaining loops not concerning compositions.
Three potential manipulated variables are considered in the studies of controllability for the top composition, namely the reflux flow L, the distillate flow D or the reflux ratio L/D. Three potential manipulated variables are considered for the bottom purity as well, namely the boilup V, bottom flow B and the boilup ratio V/B. If all combinations are considered this leaves nine possible combinations for pairing. In order to assess the various control structures, the dynamic Bristol's Relative Gain Array method (RGA) and Singular Value Analysis (SVA) have been employed. Both methods are based on linear models and are described by e.g. Skogestad and Postlethwaite (1996). For this purpose the full order model has been linearized at the stationary operation point, and represented as a transfer function model:

$$\mathbf{y}(s) = \mathbf{G}(s)\mathbf{u}(s) \tag{1}$$

Where $\mathbf{G}(s)$ is the process transfer function matrix in the Laplace domain, linking the inputs, $\mathbf{u}(s)$, to the outputs, $\mathbf{y}(s) = (Y_D, X_B)^T$. Logarithmic compositions, Y_D and X_B, for the outputs have been applied as suggested by Skogestad and Morari (1988):

$$Y_D = \ln(1 - y_D) \tag{2}$$

$$X_B = \ln x_B \tag{3}$$

Based on this transfer function matrix the frequency dependent RGA is given by:

$$\mathbf{\Lambda}(j\omega) = \mathbf{G}(j\omega) \otimes [\mathbf{G}(j\omega)^{-1}]^T \tag{4}$$

The diagonal elements in a 2×2 RGA, $\mathbf{\Lambda}$, are identical and will be referred to as λ. The operator "$\otimes$" is the Schur product, i.e. the element-wise multiplication operator.

In SVA the process transfer function matrix is decomposed to orthonormal matrices, $\mathbf{Z}$ and $\mathbf{V}$, and a diagonal matrix $\mathbf{S}$, according to:

$$\mathbf{G}(s) = \mathbf{Z}(s)\mathbf{S}(s)\mathbf{V}(s)^T \tag{5}$$

The condition number of the process transfer function matrix is given by the ratio between the largest and the smallest, non-zero diagonal element (or singular value) in $\mathbf{S}$:

$$\gamma(j\omega) = \frac{\sigma^*(j\omega)}{\sigma_*(j\omega)} \tag{6}$$

When applying SVA, scaling of input and output variables must be applied. For this purpose the flow input variables are scaled with the valve gain equal to twice the magnitudes of the nominal values while ratios are simply scaled by their nominal values.

The magnitude of the relative gain element, λ, relates to the degree of interaction between the inputs and outputs. The desired value of the relative gain element is unity. This corresponds to the case where two single loops can be designed independently since no loop interaction exists. The condition number quantifies the sensitivity of the system to uncertainties in the matrix. Physically this means that the gain of the plant depends strongly on the input direction. A high condition number indicates poor conditioning of the process matrix, thus a high condition number indicate that a process is difficult to control. A value of 10 and above is typically considered as the high-end condition number of the process matrix. The frequency range of importance for feedback control in a distillation column, is the range of 0.01-1 min^{-1} corresponding to 0.6-60 h^{-1} (Skogestad et al., 1990).

3.3 Results and Discussion

This section provides a comparison between a HIDiC and a CDiC. The steady-state RGA elements and condition numbers for the CDiC and the HIDiC are listed in Table 3 with the given control structures. The dynamic RGA

elements and condition numbers are illustrated in Figure 3. When considering the steady-state values in Table 3 the condition number varies significantly between the control structures. It is clear that the LB- and $(L/D)(V/B)$-structures are the best candidates for both distillation column configurations. No information is available for the DB-structure due to open-loop instability. Typically this steady-state analysis does not provide the complete picture since e.g. DB is a feasible structure when the loop is closed. When it comes to the dynamic analysis it can be seen in Figure 3 that the HIDiC has a greater degree of interaction than its conventional counterpart, since roughly all the lines representing the HIDiC have moved further away from unity compared to the lines belonging to the CDiC. Note the RGA element converges to unity (i.e. completely uncoupled system) at high frequencies since a hydraulic lag exist within the column. An interesting observation is the significant influence on the control structures involving ratios, $L(V/B)$, $(L/D)V$ and $(L/D)(V/B)$ control structures in particular, whose coupling have increased compared to the CDiC. This is expected since composition changes causes changes in the vapor and liquid flows within the column. It seems that the LB-structure is still superior for both configurations.

The performance of the LB-structure has been evaluated using closed-loop simulation for a feedback SISO-controller. For this evaluation, a PI-controller was incorporated in the model in discrete time with a sampling time of 0.0833 h. The controller uses the reflux flow rate to control the top purity (loop $dY_D - dL$) and the bottom product flow rate to control the bottom purity (loop $dX_B - dB$). Again, logarithmic compositions have been used for controlled and measured variables. The dynamic responses of a +10% step-change in feed flow rate and a -10% step-change in feed composition is given in Figure 4. The simulations reveal a satisfactory control performance to the relative large step-changes.

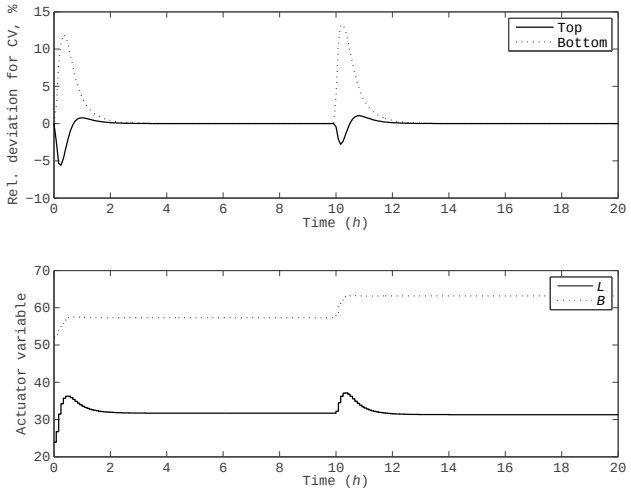

Fig. 4. Closed-loop response for the HIDiC to a +10% step-change in feed flow rate at time $t = 0$h and a -10% step-change in feed composition at $t = 15$h. Tuning parameters: $dY_D - dL$, $K_c = -0.0790$, $\tau_I = 0.5158$h and $dX_B - dB$, $K_c = 8.3147$, $\tau_I = 0.4357$h.

4. CONCLUDING REMARKS

In this paper, a complete dynamic model has been presented and utilized for dynamic analysis and simulation of a heat-integrated distillation column (HIDiC) and a conventional distillation column. Dynamic analysis showed an increased input-output coupling due to diabatization, and that otherwise feasible control structures for a conventional distillation column could turn infeasible in a HIDiC. In this study an LB-structure seemed to be a promising control structure for a HIDiC as well as a conventional distillation column, considering a case-study of a benzene-toluene separation with 99% benzene in the distillate and 5% in the bottom product.

5. NOMENCLATURE

Symbols

B	$\text{kmol} \cdot \text{h}^{-1}$	Bottom flow
c	-	Benzene Antoine parameters
D	$\text{kmol} \cdot \text{h}^{-1}$	Distillate flow
F	$\text{kmol} \cdot \text{h}^{-1}$	Feed flow
$\mathbf{G}$	-	Process transfer function matrix
j	-	Imaginary number
L	$\text{kmol} \cdot \text{h}^{-1}$	Liquid flow
M	kmol	Holdup
N	-	Number
P	MPa	Pressure
q	-	Feed thermal condition
Q	$\text{kJ} \cdot \text{h}^{-1}$	Stage heat transfer
T	K	Temperature
UA	$\text{kJ} \cdot \text{h}^{-1} \cdot \text{K}^{-1}$	Heat transfer rate
$\mathbf{u}$	-	Model input matrix
$\mathbf{U}$	-	Orthonormal basis in SVA
V	$\text{kmol} \cdot \text{h}^{-1}$	Vapor flow
x	-	Liquid light component mole fraction
X	-	Logarithmic mole fraction
y	-	Vapor light component mole fraction
Y	-	Logarithmic mole fraction
$\mathbf{y}$	-	Model output matrix
z	-	Feed light component mole fraction
$\mathbf{Z}$	-	Orthonormal basis in SVA
α	-	Relative volatility
γ	-	Condition number
λ	$\text{kJ} \cdot \text{kmol}^{-1}$	Heat transfer rate
λ	-	RGA diagonal element
$\mathbf{\Lambda}$	-	RGA
τ	h	Hydraulic time constant
σ	-	Singular value
ω	h^{-1}	Frequency

Super- and subscripts

B	Bottom
C	Condenser
D	Distillate
F	Feed
i	Stage number
R	Reboiler
s	Nominal operation condition
S	Stage

Acronyms

CDiC	Conventional distillation column
HIDiC	Heat-integrated distillation column
RGA	Relative gain array
SVA	Singular value analysis

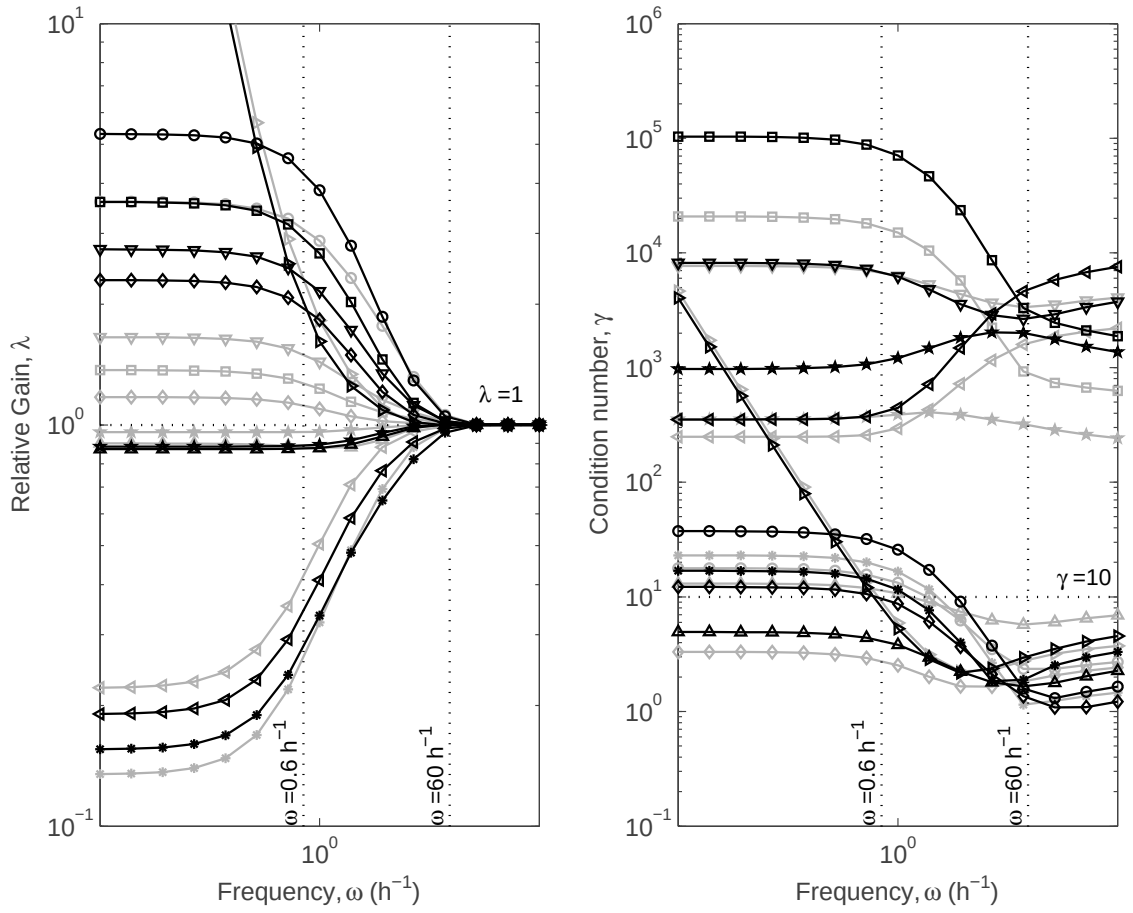

Fig. 3. Dynamic RGA Analysis and SVA applied on a HIDiC (black lines) and a CDiC (gray lines). The pressure difference between sections in the HIDiC is kept constant. **Legends:** Dotted: Lines of interest. ($\circ$): LV. ($*$): DV. ($\square$): $(L/D)V$. ($\triangle$): LB. ($\triangledown$): $L(V/B)$. ($\triangleright$): DB. ($\triangleleft$): $D(V/B)$. ($\star$): $(L/D)B$. ($\diamond$): $(L/D)(V/B)$.

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Table 1. Equations for a dynamic model being capable of describing the CDiC and the HIDiC configurations. The tray number, i, in the distillation column is counted from top to bottom. A corresponding nomenclature list is given in section 5. The operator := should be considered as a dynamic equal sign

Provide	N_S, F, z_F, q, P_i, α, L_C, V_R, $C1$, $C2$, $C3$, λ, UA and initial conditions	
Variable	Expression	Stages
Feed stage	$N_F = N_S/2 + 1$	
Vapor pressure	$P_i^* = \dfrac{P_i}{x_i + (1 - x_i)/\alpha}$	$i = 1, ..., N_S$
Temperature	$T_i = \dfrac{c_2}{c_1 - \ln P_i^*} - C_3$	$i = 1, ..., N_S$
VLE	$y_i = \dfrac{\alpha x_i}{(\alpha - 1)x_i + 1}$	$i = 1, ..., N_S$
Heat transfer	CDiC:	
	$Q_i = 0$	$i = 1, ..., N_S$
	HIDiC:	
	$Q_i = -Q_{N_F-1+i} = UA(T_{N_F-1+i} - T_i)$	$i = 1, ..., N_F - 1$
Vapor flow	$V_i = V_R + \sum_{k=i}^{N_S} Q_k/\lambda$	$i = 1, ..., N_S$
	$V_i := V_i + F(q - 1)$	$i = N_F, ..., N_S$
Liquid flow	$L_i = L_{s,i} + (M_i - M_{s,i})/\tau$	$i = 1, ..., N_S$
	Where	
	$L_{s,i} = L_{C,s} - \sum_{k=1}^{i} Q_k/\lambda$	$i = 1, ..., N_S$
	$L_{s,i} := L_{s,i} + Fq$	$i = N_F, ..., N_S$
	HIDiC:	
	$L_{N_F-1} = V_{N_F} + L_{N_F-2} - V_{N_F-1}$	
Total holdups	$dM_1/dt = V_2 + L_C - V_1 - L_1$	$i = 1$
	$dM_i/dt = V_{i+1} + L_{i-1} - V_i - L_i$	$i = 2, ..., N_S - 1$
	$dM_{N_S}/dt = V_R + L_{N_S-1} - V_{N_S} - L_i$	$i = N_S$
	$dM_{N_F}/dt := dM_{N_F}/dt + F$	$i = N_F$
Component holdup	$dx_1/dt = (V_2y_2 + L_Cx_C - V_1y_1 - L_1x_i - x_1dM_1/dt)/M_1$	$i = 1$
	$dx_i/dt = (V_{i+1}y_{i+1} + L_{i-1}x_{i-1} - V_iy_i - L_ix_i - x_idM_i/dt)/M_i$	$i = 2, ..., N_S$
	$dx_{N_S}/dt = (V_Ry_R + L_{N_S-1}x_{N_S-1} - V_{N_S}y_{N_S} - L_ix_{N_S} - x_{N_S}dM_{N_S}/dt)/M_{N_S}$	$i = N_S$
	$dx_{N_F}/dt := dx_{N_F}/dt + F(z_F - x_{N_F})/M_{N_F}$	$i = N_F$
Condenser	$dx_C/dt = (V_1y_1 - L_Cx_C - Dx_D)/M_C$	
	Total: $y_D = x_C = y_1$	
Reboiler	$dx_R/dt = (L_{N_S}x_{N_S} - V_Ry_R - Bx_R)/M_R$	
	Total: $x_R = y_R = x_B$	

Table 3. Stationary RGA elements and condition numbers for potential configurations of the CDiC and the HIDiC related to the separation of an equimolar benzene-toluene mixture

	HIDiC		CDiC	
Control structure	RGA element, λ	Condition number, γ	RGA element, λ	Condition number, γ
LV	5.31	37.43	3.60	17.77
DV	0.16	16.92	0.13	23.06
$(L/D)V$	3.60	103575	1.37	20816
LB	0.87	4.94	0.90	13.08
$L(V/B)$	2.74	8188	1.65	7701
DB	∞	∞	∞	∞
$D(V/B)$	0.19	352	0.22	248
$(L/D)B$	0.88	972	0.96	354
$(L/D)(V/B)$	2.30	12.22	1.17	3.32